



## VERIFICATION OF A TRANSLATION

I, Yanagawa Yasuo

of Mitsuya-Yotsuya Building 8th Floor, 2-14, Yotsuya,  
Shinjuku-ku, Tokyo, 160-0004 Japan

declare as follows:

1. That I am well acquainted with both the English and Japanese languages
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Inventor(s):  
Address: Heights Imura 203, Omaezaki 2386,  
Omaezaki-cho, Haibara-gun, Shizuoka,  
421-0601 Japan  
Name: Himiko Takayama  
Address: Limpia Nakayari 306, Hosoe 990-1,  
Haibara-cho, Haibara-gun, Shizuoka,  
421-0421 Japan  
Name: Katsumi Umehara

Applicant(s):  
Registration Number; 391050525  
Name; ORONITE JAPAN LIMITED

Agent:  
Registration Number; 100074675  
Patent Attorney  
Name; Yasuo Yanagawa

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S P E C I F I C A T I O N

[Title of the Invention]

Lubricating oil composition and additive composition

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[Scope of Patent Claim]

1. A lubricating oil composition comprising a base oil, in which dissolved or dispersed are at least the following components:

10 (A) 0.1 to 5.0 wt.% of at least one compound selected from the group consisting of a phosphoric acid ester, a thiophosphoric acid ester, and amine salts thereof;

(B) 0.01 to 1.0 wt.% of a phosphorous acid ester  
15 and/or an amine salt thereof;  
and

(C) 0.01 to 2.0 wt.% of at least one compound selected from the group consisting of an alkenyl succinimide, an alkenyl succinic acid ester, benzylamine, and  
20 derivatives thereof.

2. The lubricating oil composition of claim 1, in which a weight ratio of components (A):(B) is in the range of 1:1 to 500:1.

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3. The lubricating oil composition of claim 1, in which a weight ratio of components (B):(C) is in the range of 1:0.5 to 1:20.

4. The lubricating oil composition of claim 1, in which a total phosphorus content is in the range of 50 to 5,000 mass ppm.

5 5. The lubricating oil composition of claim 1, in which the base oil has a kinematic viscosity of 5 to 900 mm<sup>2</sup>/s at 40°C.

6. The lubricating oil composition of claim 1,  
10 which is to be used as a hydraulic oil, a bearing oil, an industrial gear oil, or a sliding surface lubricating oil.

7. A lubricating oil additive composition compris-  
15 ing a base oil, in which dissolved or dispersed are at least the following components:

(A) 10 to 90 wt.% of at least one compound selected from the group consisting of a phosphoric acid ester, a thiophosphoric acid ester, and amine salts thereof;

20 (B) 1 to 20 wt.% of a phosphorous acid ester and/or an amine salt thereof;

and

(C) 1 to 40 wt.% of at least one compound selected from the group consisting of an alkenyl succinimide, an  
25 alkenyl succinic acid ester, benzylamine, and derivatives thereof.

8. The lubricating oil additive composition of claim 7, in which a weight ratio of components (A):(B) is  
30 in the range of 1:1 to 500:1.

9. The lubricating oil additive composition of claim 7, in which a weight ratio of components (B):(C) is in the range of 1:0.5 to 1:20.

5

10. The lubricating oil additive composition of claim 7, in which a total phosphorus content is in the range of 0.5 to 20 wt.%.

10 11. The lubricating oil additive composition of claim 7, which is to be used for preparing a hydraulic oil, a bearing oil, an industrial gear oil, or a sliding surface lubricating oil.

15 [Detailed Description of the Invention]

Field of Invention

The present invention relates to a lubricating oil composition which shows excellent heat-resistant and anti-wear characteristics at a high temperature and a  
20 high pressure which is favorably employable as lubricating oil other than internal combustion engine lubricating oil, particularly a hydraulic oil, and further relates to an additive composition which is favorably employable for the preparation of the lubricating oil.

25

Prior Art

Heretofore, in a hydraulic oil, a zinc dithiophosphate (ZnDTP) has been employed as an additive. While the zinc dithiophosphate is excellent in anti-wear and  
30 oxidation-inhibition properties, its heat-resistance is

not satisfactorily high. At present and in future in which working conditions for hydraulic oil get more severe, it may decompose by heat and produce sludge.

Therefore, in recent apparatuses exposed to high temperatures, an ashless hydraulic lubricating oil in which a phosphorus extreme pressure agent (which shows high thermal stability) is contained is generally used in place of the conventional zinc-containing hydraulic oil. However, an ashless hydraulic oil containing the phosphorus extreme pressure agent is relatively poor in its extreme pressure resistance and anti-wear performance, as compared with the zinc-containing hydraulic oil. Therefore, the phosphorus extreme pressure agent-containing hydraulic oil cannot always be employed widely.

It is required, for a bearing oil, an industrial gear oil, and a sliding surface-lubricating oil, extreme pressure resistance and anti-wear performance higher than those required for a hydraulic oil. Therefore, in these lubricating oils, a phosphorus-containing extreme pressure agent and a sulfur-containing extreme pressure agent are employed in combination. However, a lubricating oil having high extreme pressure resistance is apt to show poor thermal stability and sometimes produces sludge.

Therefore, it is not easy to produce a lubricating oil showing both of enough high extreme pressure resistance and enough anti-wear performance.

Japanese Patent Provisional Publication No. 9-111277 describes an ashless hydraulic oil composition comprising %C<sub>A</sub>5 or less of a base oil, (A) 0.01-5 wt.% of an amine-type oxidation inhibitor, (B) 0.01-5 wt.% of a phenolic

oxidation inhibitor, (C) 0.01-5 wt.% of a phosphoric acid ester, and (D) 0.001-5 wt.% of an aliphatic amide and/or a polyhydric alcohol ester.

Japanese Patent Provisional Publication No. 11-  
5 323365 describes a hydraulic oil using a mineral oil, a synthetic oil, or a mixture thereof, as a base oil, which further contains (A) 0.01-1 wt.% of an alkenyl succinimide or its derivative, (B) 0.1-5 wt.% of a phosphoric acid ester, (C) 0.05-0.5 wt.% of an alkylated diphenyl-  
10 amine, and (D) 0.05-0.5 wt.% of a hindered phenol.

#### Problems to be Solved by Invention

It is an object of the invention to provide a lubricating oil which shows high thermal stability and high  
15 extreme pressure resistance and which gives sludge in less amount, even at a high temperature and a high pressure and which is favorably employed for lubricating hydraulic systems, bearing area, industrial gear, and sliding surface.

20 It is another object of the invention to provide an additive composition which is favorably employable for preparation of the above-mentioned lubricating oil.

#### Means to solve Problems

25 The present invention resides in a lubricating oil composition comprising a base oil, in which dissolved or dispersed are at least the following components:

(A) 0.1 to 5.0 wt.% of at least one compound selected from the group consisting of a phosphoric acid

ester, a thiophosphoric acid ester, and amine salts thereof;

(B) 0.01 to 1.0 wt.% of a phosphorous acid ester and/or an amine salt thereof;

5 and

(C) 0.01 to 2.0 wt.% of at least one compound selected from the group consisting of an alkenyl succinimide, an alkenyl succinic acid ester, benzylamine, and derivatives thereof.

10 The invention further resides in a lubricating oil additive composition comprising a base oil, in which dissolved or dispersed are at least the following components:

(A) 10 to 90 wt.% of at least one compound selected  
15 from the group consisting of a phosphoric acid ester, a thiophosphoric acid ester, and amine salts thereof;

(B) 1 to 20 wt.% of a phosphorous acid ester and/or an amine salt thereof;

and

20 (C) 1 to 40 wt.% of at least one compound selected from the group consisting of an alkenyl succinimide, an alkenyl succinic acid ester, benzylamine, and derivatives thereof.

#### 25 Embodiment of the Invention

The lubricating oil composition of the invention comprises a mineral or synthetic base oil having a kinematic viscosity of 5 to 900 mm<sup>2</sup>/s, preferably 20 to 700 mm<sup>2</sup>/s at 40°C. A mineral oil employable for the invention  
30 can be obtained from crude oil by distillation (under



atmospheric or reduced pressure) and purification such as solvent extraction, hydrocracking, solvent dewaxing or hydrogenation refining. Particularly preferred is a highly hydrogenation-refined base oil having a viscosity  
5 index of 100 to 150, an aromatic content of 5 wt.% or less, a nitrogen content of 50 ppm or less, and a sulfur content of 50 ppm or less.

The synthetic oil (i.e., synthetic lubricating base oil) can be poly- $\alpha$ -olefin which is a polymer of  $\alpha$ -olefin  
10 having 3 to 12 carbon atoms; a dialkyl diester such as dioctyl sebacate, which is an ester of a dibasic acid (e.g., sebacic acid, azelaic acid, or adipic acid) and an alcohol having 4 to 12 carbon atoms; a polyol ester which is an ester of a monobasic acid having 3 to 18 carbon  
15 atoms and 1-trimethylolpropane or pentaerythritol, or an alkylbenzene having an alkyl group which contains 9 to 40 carbon atoms.

The mineral oil and synthetic oil can be employed singly. If desired, two or more of mineral oils and two  
20 or more of synthetic oils can be employed in combination.

If desired, a mineral oil and a synthetic oil can be employed in combination with an optionally chosen ratio.

The lubricating oil of the invention contains 0.1 to 5.0 wt.%, preferably 0.1 to 3 wt.%, of (A) at least one  
25 compound selected from the group consisting of a phosphoric acid ester, a thiophosphoric acid ester, an amine salt thereof. The amount means a ratio based on the total amount of the lubricating oil, and each component of the indicated amount contains a small amount of hydro-

carbon oil which is employed in the preparation of the component and remains in the component.

The phosphoric acid ester, thiophosphoric acid ester, and amine salt thereof functions to enhance the lubricating performances, and can be selected from known compounds which *per se* have been conventionally employed as extreme pressure agents. Generally employed are a phosphoric acid ester, a thiophosphoric acid ester, or an amine salt thereof which has an alkyl group, an alkenyl group, an alkylaryl group, or an aralkyl group, any of which contains approximately 3 to 30 carbon atoms.

Examples of the phosphoric acid esters include aliphatic phosphoric acid esters such as triisopropyl phosphate, tributyl phosphate, ethyl dibutyl phosphate, trihexyl phosphate, tri-2-ethylhexyl phosphate, trilauryl phosphate, tristearyl phosphate, and trioleyl phosphate; and aromatic phosphoric acid esters such as benzyl phenyl phosphate, allyl diphenyl phosphate, triphenyl phosphate, tricresyl phosphate, ethyl diphenyl phosphate, cresyl diphenyl phosphate, dicresyl phenyl phosphate, ethylphenyl diphenyl phosphate, diethylphenyl phenyl phosphate, propylphenyl diphenyl phosphate, dipropylphenyl phenyl phosphate, triethylphenyl phosphate, tripropylphenyl phosphate, butylphenyl diphenyl phosphate, dibutylphenyl phenyl phosphate, and tributylphenyl phenyl phosphate.

Examples of the thiophosphoric acid esters include aliphatic thiophosphoric acid esters such as triisopropyl thiophosphate, tributyl thiophosphate, ethyl dibutyl thiophosphate, trihexyl thiophosphate, tri-2-ethylhexyl

thiophosphate, trilauryl thiophosphate, tristearyl thiophosphate, and trioleyl thiophosphate; and aromatic thiophosphoric acid esters such as benzyl phenyl thiophosphate, allyl diphenyl thiophosphate, triphenyl thiophosphate, tricresyl thiophosphate, ethyl diphenyl thiophosphate, cresyl diphenyl thiophosphate, dicresyl phenyl thiophosphate, ethylphenyl diphenyl thiophosphate, diethylphenyl phenyl thiophosphate, propylphenyl diphenyl thiophosphate, dipropylphenyl phenyl thiophosphate, triethylphenyl thiophosphate, tripropylphenyl thiophosphate, butylphenyl diphenyl thiophosphate, dibutylphenyl phenyl thiophosphate, and tributylphenyl thiophosphate.

Also employable are amine salts of the above-mentioned phosphates and thiophosphates. Also employable are amine salts of acidic alkyl or aryl esters of the phosphoric acid and thiophosphoric acid.

The components (A) can be employed singly or in combination.

If the content of the phosphoric acid ester, thiophosphoric acid ester, and/or their amine salts in the lubricating oil is less than 0.1 wt.%, enough lubricating performance cannot be brought about into the oil. Even if the content is more than 5.0 wt.%, no further improvement is expected, and thus such content is disadvantageous from the economic viewpoint.

The lubricating oil of the invention further contains 0.01 to 1.0 wt.%, preferably 0.01 to 0.4 wt.% of (B) a phosphorous acid ester and/or an amine salt thereof. The amount means a ratio based on the total amount of the lubricating oil, and each component of the indi-

cated amount contains a small amount of hydrocarbon oil which is employed in the preparation of the component and remains in the component.

In the lubricating oil of the invention, the component (A) and component (B) are preferably employed in a (A):(B) weight ratio of 1:1 to 500:1, more preferably 1:1 to 50:1.

The phosphorous acid ester and/or its amine salt function to enhance the lubricating performances, and can be selected from known compounds which *per se* have been conventionally employed as extreme pressure agents. Generally employed are a phosphorous acid ester, or an amine salt thereof which has an alkyl group, an alkenyl group, an alkylaryl group, or an aralkyl group, any of which contains approximately 3 to 30 carbon atoms.

Examples of the phosphorous acid esters include aliphatic phosphorous acid esters such as triisopropyl phosphite, tributyl phosphite, ethyl dibutyl phosphite, trihexyl phosphite, tri-2-ethylhexylphosphite, trilauryl phosphite, tristearyl phosphite, and trioleyl phosphite; and aromatic phosphorous acid esters such as benzyl phenyl phosphite, allyl diphenylphosphite, triphenyl phosphite, tricresyl phosphite, ethyl diphenyl phosphite, tributyl phosphite, ethyl dibutyl phosphite, cresyl diphenyl phosphite, dicresyl phenyl phosphite, ethylphenyl diphenyl phosphite, diethylphenyl phenyl phosphite, propylphenyl diphenyl phosphite, dipropylphenyl phenyl phosphite, triethylphenyl phosphite, tripropylphenyl phosphite, butylphenyl diphenyl phosphite, dibutylphenyl phenyl phosphite, and tributylphenyl

phosphite. Also favorably employed are dilauryl phosphite, dioleoyl phosphite, dialkyl phosphites, and diphenyl phosphite.

Amine salts of these phosphorous acid esters are also employable. The component (B) can be used singly or in combination.

If the content of the phosphorous acid ester, and/or its amine salt in the lubricating oil is less than 0.01 wt.%, enough lubricating performance cannot be brought about into the oil. Even if the content is more than 1.0 wt.%, no further improvement is expected, and thus such content is disadvantageous from the economic viewpoint.

The lubricating oil of the invention further contains 0.01 to 2.0 wt.%, preferably 0.01 to 1.0 wt.% of (C) a compound selected from the group consisting of an alkenyl succinimide, an alkenyl succinic acid ester, benzyl amine, and derivatives thereof. The amount means a ratio based on the total amount of the lubricating oil, and each component of the indicated amount contains a small amount of hydrocarbon oil which is employed in the preparation of the component and remains in the component.

In the lubricating oil of the invention, the component (B) and component (C) are preferably employed in a (B):(C) weight ratio of 1:0.5 to 1:20, more preferably 1:1 to 1:3.

The alkenyl succinimide can be a monoimide or a bisimide, and can be prepared by reaction between a polybutenyl succinic anhydride and a polyamine. The polybutenyl succinic anhydride can be produced by reaction of

a polybutene having a mean molecular weight of 800 to 8,000 or a chlorinated polybutene having a mean molecular weight of 800 to 8,000 with maleic anhydride at a temperature of 100 to 200°C. Examples of the polyamines  
5 include diethylene triamine, triethylene tetramine, tetraethylene pentamine, pentaethylene hexamine, and hexamethylene heptamine.

Examples of the alkenyl succinimide derivatives include a borated derivative, an organic phosphonate  
10 derivative, and a derivative which is produced by reacting an alkenyl succinimide with aldehyde, ketone, carboxylic acid, sulfonic acid, alkylene oxide, sulfur, or polyhydric alcohol. A preferred derivative is a borated derivative, which can be produced by reacting the poly-  
15 butenyl succinic anhydride-polyamine reaction product with boric acid or a boric acid derivative.

The alkenyl succinic acid ester and its derivative can be an ester of the above-mentioned polybutenyl succinic acid which has been prepared by the reaction be-  
20 tween a polybutene or a chlorinated polybutene and maleic anhydride, with a polyhydric alcohol such as pentaerythritol, and its derivative.

The benzylamine and its derivative can be prepared by reacting the above-mentioned polybutene with phenol,  
25 formaldehyde and polyamine.

The component (C) can be used singly or in combination.

If the content of the component (C) in the lubricating oil is less than 0.01 wt.%, enough sludge dispersing  
30 performance cannot be brought about into the oil and

further poor water tolerance is given. If the content is more than 2.0 wt.%, oxidation stability likely decreases.

The lubricating oil composition can further contain a variety of known additives, such as oxidation inhibitors, metal-inactivating agents, demulsifiers, anti-foaming agents, and pour point depressants.

The lubricating oil composition of the invention can be prepared by successively or simultaneously adding the aforementioned components to a base oil, or by beforehand preparing the additive composition in several ten-fold to one hundred-fold concentration and then mixing it with a base oil.

#### Examples

[Examples 1 to 3]

The following base oil and additive components were blended in amounts (wt.%) set forth in Table 1, to prepare lubricating oil compositions. The total phosphorus contents (mass ppm) were those set forth in Table 1:

- (1) base oil: petroleum-origin,  
viscosity (at 40°C) approx. 36 cSt
- (2) tricresyl phosphate solution: TCP (available from Daihachi Chemicals Co., Ltd.)
- (3) triphenyl thiophosphate and its derivative solution  
(Irgalube 232, available from Ciba Speciality Chemicals, Inc.)
- (4) dilauryl phosphite solution (JP 212, Johoku Chemicals Co., Ltd.)
- (5) alkenylsuccinimide (OLOA 371, available from Oronite Japan Co., Ltd.)

- (6) auxiliary additives (oxidation inhibitor, metal-inactivating agent, demulsifier, anti-foaming agent, etc.)

5 [Comparison Examples 1 to 5]

The above-mentioned additive components were blended to prepare lubricating oil compositions.

[Evaluation of Lubricating Oil Compositions]

10 The lubricating oil compositions of Examples and Comparison Examples were evaluated in their lubricating performances by the following tests. The results of the tests are set forth in Table 1.

(1) CM thermal stability test

15 The test oil (lubricating oil composition) was heated under the following conditions:

Test temperature: 150°C

Test period: 168 hours.

The test oil was then filtered over a filter (pore size: 0.8  $\mu$ m), washed with n-hexane, and dried. The dry residue on the filter was weighed to determine the amount of sludge.

(2) Shell four-ball test

20 The test oil was placed in a Shell four-ball tester, which was then operated at 1,800 r.p.m., to determine an initial seizure load (ISL).



Table 1-(1)

		(1)	(2)	(3)	(4)	(5)	(6)	P (ppm)
5								
	Ex. 1	99.425	0.200	-	0.025	0.050	0.300	180
	Ex. 2	98.850	0.400	-	0.050	0.100	0.600	360
10	Ex. 3	99.425	-	0.200	0.025	0.050	0.300	160
	Com.1	99.500	0.200	-	-	-	0.300	160
	Com.2	99.450	0.200	-	-	0.050	0.300	160
	Com.3	99.475	0.200	-	0.025	-	0.300	180
15	Com.4	99.675	-	-	0.025	-	0.300	20
	Com.5	99.625	-	-	0.025	0.050	0.300	20

Table 1-(2)

Table 1 (2)

20	CM thermal stability test		Shell 4-ball test	
	Sludge amount	Viscosity	ISL	
	(mg)	increase (%)	(kg)	
25				
	Example 1	1.0	5	80
	Example 2	1.4	2	100
	Example 3	1.0	0	80
30				
	Com. Ex. 1	0.8	5	63
	Com. Ex. 2	1.0	7	63
	Com. Ex. 3	1.9	6	80
35	Com. Ex. 4	1.9	8	80
	Com. Ex. 5	0.5	10	63

As is apparent from the results set forth in Table 1, in the test conditions of a high temperature and a long test period, the lubricating oil compositions of the invention give sludge and viscosity increase less than those given by the conventional lubricating oil compositions. Further, the lubricating oil compositions of the invention show a high initial seizure load, which means excellent extreme pressure resistance.

10

#### Effect of the Invention

The lubricating oil composition of the invention shows a high thermal stability and a high extreme pressure resistance under the conditions of a high temperature and a high pressure. Further, the lubricating oil composition of the invention efficiently reduces production of sludge for a long period of time. Thus, the lubricating oil composition of the invention shows excellent performance for the use as a lubricating oil other than internal combustion engine lubricating oil, and therefore the lubricating oil composition of the invention is favorably employable, specifically, for lubricating hydraulic systems, bearing systems, industrial gear systems, and sliding systems.

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A B S T R A C T

[Problem]

5        To provide a lubricating oil composition which shows  
high thermal stability and high extre pressure resistance  
and effectively reduce production of sludge under the  
conditions of a high temperatures and an extreme pres-  
sure, so that the oil composition is favorably employable  
10 for lubricating hydraulic systems, bearing systems,  
industrial gear systems, and sliding systems.

[Means to Solve the Problem]

15        A lubricating oil composition comprising a base oil  
and additives dissolved or dispersed in the base oil, in  
which the additives are 0.1 to 5.0 wt.% of (A) phosphoric  
acid ester, thiophosphoric acid ester or its amine salt;  
0.01 to 1.0 wt.% of (B) phosphorous acid ester or its  
amine salt; and 0.01 to 2.0 wt.% of (C) alkenyl succin-  
20 imide, alkenyl succinic acid ester, benzylamine or its  
derivative.

[Selected Drawings]

None